Biomass Gasification Polygeneration

Designing and operating two-stage systems coupled with solid oxide cell technology

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Publication date:
2018

Document Version
Publisher's PDF, also known as Version of record

DTU Chemical Engineering
Department of Chemical and Biochemical Engineering

BIOMASS GASIFICATION
POLYGENERATION

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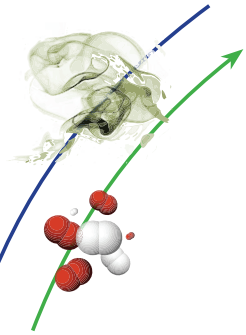


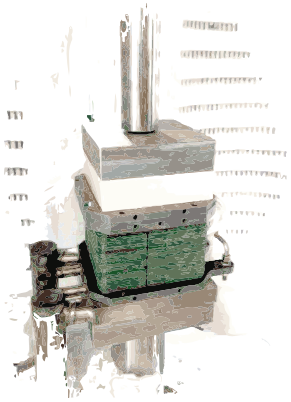


Ph.D. thesis by
Rasmus Østergaard Gadsbøll

June, 2018

Preface and acknowledgement

This thesis is submitted as a partial fulfilment of the requirements for the PhD degree at the Technical University of Denmark.

The project was carried out as a part of the Biomass Gasification Polygeneration research project, ForskVE-12205 (transferred to EUDP during the project), at Energinet over a 3-year study at the Department of Chemical and Biochemical Engineering, Section of Combustion and Harmful Emission Control (CHEC) from December 2014 to June 2018. The main supervisor senior researcher Ulrik Birk Henriksen (CHEC) was accompanied by co-supervisors senior researcher Jesper Ahrenfeldt (CHEC) and associate professor Lasse Røngaard Clausen from the Department of Mechanical Engineering. Project partners include Danish Gas Technology Center and Dall Energy.

First and foremost, I would like to thank my supervisors Ulrik, Jesper and Lasse for their support and interest during the project. The very informal, open minded and straight forward tone during discussions and reviews, have really allowed me to think independently and creatively, and allowed for continuous development of the project. This environment and process has really molded and given direction to the final products.

I would also like to thank Ulrik and Jesper as daily leaders of the research group, as well as my excellent colleagues(!) at the Biomass Gasification Group. I really credit all of you for constructing and supporting a work environment that relies on empowerment, flexibility and positive attitude. These settings have really helped me during my studies to spark creative and critical thinking as well as helping me to develop my personal skills and keep my motivation high.

Thanks!

_____

Rasmus Østergaard Gadsbøll

DTU Risø, June 2018

Response to the preliminary assessment of the PhD study

This is a slightly revised version of the Ph.D. thesis. Several suggestions for minor corrections have been made by the PhD assessment committee members. Besides very minor (typos etc.) corrections, three changes have been implemented and they are listed here as committees statements (black) and my responses (red).

- The synopsis (chapters 1, 2 and 8) is quite short. One reason is the decision of not including a complete literature survey in these. It is suggested to expand sections 1.4 Methodology and/or 1.5 Thesis outline to explain more about the approach, the content of the individual studies and the connection between these.
    - As suggested, I have detailed the Methods section 1.4 (previously Methodology) by formulating a paragraph on how the methods relate to the thesis and each other: "*These steps has been completed in nearly chronological order as stated above. The methods have been formulated in five scientific articles and a patent, which constitutes the core of the work for this PhD project. Each of the articles represent individual pieces of research and hence contain their own literature survey, framing, analysis and results. This thesis then acts as a vehicle for connecting these publications by framing them in a larger context in the start (PhD formalia, project introduction and project technical background) and in the end (summarizing, analyzing and discussing across the studies and in a larger energy system perspective related to the initial project framing and subsequent developments).*"

- Page VIII: The purpose of the section entitled “Teaching” is not clear. It may be removed or clarified for example changing the title to, e.g., “Student supervision” and adding student and supervisor names.
    - Teaching activities have been clarified. The activities constitutes student supervision and mentoring during experimental projects.

- Page 13: A statement about “limits of the Carnot cycle” is included without reference or further explanation. This seems to be based on simplifications that are not aligned with level of the thesis as such. The paragraph may be clarified.
    - I agree that the commnent on the Carnot efficiency is not sufficiently well-written and I have made a reformulation of the paragraph: "*The fuel cell utilizes separate flows of fuel and oxidizer and an ion-conducting membrane to produce electricity directly via electrochemical reactions. Because of this characteristic it is not subject to the limits of the Carnot cycle as traditional heat engines, but instead traditionally[1] to the limits of the relationship between the change in Gibbs free energy (the released electrical energy) and the change in enthalpy of formation (the calorific value). While this does not make the fuel cell more efficient than heat engines in all cases, this mechanism enables the fuel cell to operate at very high thermodynamic efficiency that typically outperforms that of heat engines.*

The response was sent to the committee on the 20th of November 2018, two weeks prior to the Ph.D. defence.

Summary

Because of the increasing implementation of fluctuating wind and solar energy sources, there is a need to further integrate the power, heat and transport sectors. This can be done via implementation of biomass-based flexible energy plants that can alter operation based on the current state of the grids and convert power into effective and easily storable energy carriers. This study investigates a proposed *polygeneration* concept that can either produce or consume power, and store electricity as biofuels. The concept consist of a joint platform of thermal biomass gasification and solid oxide cell technologies that form a very efficient and flexible system. This system has two operational states: 1) The gasifier operates with air and the resulting product gas is fed to the solid oxide fuel cell (SOFC) for power production; 2) The cells are now operated as electrolysis cells (SOEC), converting power and steam into H_2 and O_2, which is then fed to the gasifier that produces a N_2-free gas suited for biofuel synthesis. The polygeneration study is centered around the state-of-the-art TwoStage gasification concept. This project seeks to provide the foundation for future polygeneration plants by performing experimental proof-of-concepts and analysis for the two operational states, and provide theoretical investigations of large-scale configurations of the gasification system. The project is divided into three main parts.

The first part of the project studies the coupling of the 80kW$_{th}$ TwoStage Viking pilot gasifier plant and an 800W$_e$ SOFC stack from Topsoe Fuel Cell for power production. The coupling has been suggested as highly efficient and competitive, and the aim of this study is therefore to assess the performance characteristics of this coupling. This was done during two experimental campaigns respectively.
Initially, the SOFC was operated with product gas, whilst applying an active carbon filter, scrubber and desulphurizer. The gas cleaning is found to be more than sufficient. The SOFC part-load performance was tested down to 55% flow rate and is found to maintain its electric efficiency. The peak electric efficiency is found to 46% gas-to-power, which is the highest reported value in the literature for product gas operation. The performance correlates to ≈40% biomass-to-power efficiency for the entire system, which is in line with previous investigations.
In the second campaign the impact of applying minimal or no gas cleaning between the gasifier and SOFC is studied along with operational characteristics. Initially, the gas cleaning train was simplified to a carbon filter and desulphurizer, and tests were performed with an air-blown product gas at SOFC operating temperatures of 700^{o}C and 800^{o}C. A power and efficiency increase of 8-11% and 4%-points respectively is found at 800^{o}C. Changing to an O_2-CO_2-blown product gas is found to effect the performance negatively. Lastly, the carbon filter and desulphurizer were bypassed, no short-term changes in operational voltage was seen with 1.5-2.8ppm sulphur in the feed gas. This indicates that the gasifier design can be a key feature when constructing gas cleaning for gasifier-SOFC systems, as in-situ gas cleaning can reduce the downstream cleaning significantly.

The second part of the project performs a comparative study of the Viking pilot plant, where the typical air-blown configuration were changed to O_2-CO_2-blown operation. The purpose is to achieve an N_2-free product gas and analyze the corresponding gasifier operation, performance and gas quality. This was initially investigated via thermodynamic modeling, where it is seen that the operational characteristics are within range of air if 21v% O_2-in-CO_2 is applied and nearly identical parameters at 30v% O_2. The pilot plant was then modified and prepared and an experimental campaign was carried out with air, 21v% O_2–in-CO_2 and 25v% O_2–in-CO_2. The results validated the tendencies of the modeling studies, as operation temperatures were seen to slightly decrease at 21v% O_2-in-CO_2: the partial oxidation temperature by 52-

69°C and grate temperature by 31-36°C. As expected, 25v% O_2 showed characteristics between these value and air-operation. Detailed gas analysis showed that the tar and sulphur concentrations of a few mg/Nm^3 and <3ppm respectively, are similarly low across the gasification media and that the N_2-content is reduced to a few percentages.

The third part of the project analyzed the possibility to develop large-scale plant designs of >10MW_{th} based on the TwoStage gasification concept and to investigate whether they could be integrated in a polygeneration context respectively. Common for the two studies were the development of a wide knowledgebase of biomass gasification subprocesses, current state-of-the-art plant and previous TwoStage upscaling developments etc., which through creative processes and thermodynamic modelling led to the results of the study.
In the initial study, the project analyses the potential for wood-based fixed bed and fluid bed systems. Amongst several, two noteworthy new design features was implemented in the concepts: 1) applying a pyrolysis reactor with recirculation of gas for utilizing available heat sources indirectly and avoid dilution; 2) applying subsequent cooling of the hot partial oxidation gases with drying steam, either as direct injection or via ejector-driven gas recirculation. This is done to enable high temperatures and match the gasification reactor and fuel tolerances. These two features led to three main design concepts, which were shown to achieve very high cold gas efficiencies and are expected to require minimal tar removal downstream. The three concepts achieved cold gas efficiencies of 88-93%. The concept with two updraft reactors proved to be especially interesting with its high efficiency of 93% and increased tolerance to fines compared to the current downdraft constellation of the TwoStage system. While still in the developmental phase, the cold gas efficiencies were found to be 6-22%-points higher than current state-of-the-art gasifiers, whilst applying minimal or very simple gas cleaning in comparison.
In the second study, further developmental work and analysis of fuel and operational flexibility were investigated. The aim was to design systems capable of polygeneration with air and 50v% O_2-H_2O. Three different plant configurations are presented: all apply fluid bed gasification reactors, but with either an updraft, slow fluid bed or fast fluid bed pyrolysis coupling. In order to expand the analysis, the use of wood and straw fuels are investigated for all concepts and operational states. The results show relatively stable plant operation and cold gas efficiencies are calculated on average to 83-88%. The concept featuring an updraft pyrolyzer proves the most promising with regards to simplicity and cold gas efficiencies. In line with the initial study, the concepts are found to have significant potential and be – on a technical basis - competitive with current state-of-the-art gasifier with regards to cold gas efficiency, gas cleaning equipment and fuel flexibility.

Polygeneration concepts have been suggested by several Danish energy entities and will likely be featured in the future energy system towards 2030-2050 in 50-200MW_{th} capacities. Based on this work it is concluded that the TwoStage-polygeneration concept is technically feasible from a gasifier-centric point of view. This study shows that the gasification system can be modified for both SOFC and oxygen-operation without extensive additions to the existing setup and change in operation. Additionally, novel, scalable TwoStage concepts has been design and modeled, and has shown rare cold gas efficiencies and operational flexibility. Hence, the proposed concept is primed for the transition towards a sustainable energy system.

Dansk sammenfatning

I forbindelse med den stadigt stigende integration af fluktuerende vind- og solenergi, er der et behov for en større integration af el-, varme- og transportsektorerne. Dette kan gøres ved at implementere biomasse-baserede fleksible energianlæg, der kan skifte driftstilstand alt efter den givne situation på nettene og konvertere el til effektive og let lagerbare energibærere. Dette studie undersøger et *polygeneration*-koncept, der enten kan producere eller forbruge el, og tilmed lagre el i form biobrændsler. Konceptet er bygget op om en koblet platform af termisk biomasse forgasnings- og fast-oxid celle-teknologi, der samlet danner et meget effektiv og fleksibelt system. Dette system har to driftstilstande: 1) Forgasseren er i drift med luft og den resulterende produktgas fødes til en fast-oxid brændselscelle (SOFC) for elproduktion; 2) Cellen sættes nu i drift som elektrolysecelle (SOEC), der konvertere el og damp til brint og ilt, der fødes forgasseren, som så kan producerer en kvælstoffri gas som er velegnet til biobrændstofsyntese. Omdrejningspunktet for studiet af polygeneration-konceptet er To-trins forgasningskonceptet. Dette projekt sigter på, at danne et grundlag for fremtidige polygeneration-anlæg ved at foretage proof-of-concept-forsøg og analyser af de to driftstilstande, samt at leverer teoretiske undersøgelser af stor-skala konfigurationer af forgasningssystemet. Projektet er opdelt i tre hoveddele.

Den første del af projektet studerer koblingen mellem 80kW$_{th}$ To-trins Viking pilotanlægget og en 800W$_e$ SOFC-stak fra Topsoe Fuel Cell til elproduktion. Koblingen er blevet peget på som høj-effektiv og konkurrencedygtigt, og målet for dette studie er derfor at kortlægge driftskarakteristika for koblingen. Dette blev undersøgt over to eksperimentelle forsøgsperioder.
Først blev SOFC stakken sat i drift med produktgas via et aktivt kulstoffilter, scrubber og afsvovler. Det konkluderes at gasrensningen er mere end tilstrækkelig. SOFC'ens dellastkarakteristika blev testet ned til 55% flow og data viser at den elektriske virkningsgrad fastholdes på tværs af flow. Den maksimale elektriske virkningsgrad er bestemt til 46% gas-til-el, hvilket er den højest rapporterede værdi i litteraturen for drift med produktgas. Systemet kan således opnå ≈40% biomasse-til-el virkningsgrad i nuværende udformning, hvilket er på linje med tidligere undersøgelser.
I den anden forsøgsperiode blev betydning af minimal eller ingen gasrens mellem forgasseren og SOFC-stacken vurderet sammen med driftskarakteristika. Først blev gasrensning simplificeret til et kulstoffilter og en afsvovler, og der blev udført test med luftblæst produktgas ved 700^{o}C og 800^{o}C. En stigning i elproduktion of -virkningsgrad på 8-11% og 4%-point henholdsvis er fundet ved 800^{o}C. Ved at skifte til en O_2-CO_2-blæst produktgas opnås lavere performance fra stacken. Til sidst blev kulstoffilteret og afsvovleren omkringgået: ingen kort-tids effekter i SOFC-spændingen ses med 1.6-2.8ppm svovl i produktgassen. Dette indikerer at forgasserdesign kan være en af nøglerne til at designe gasrensningssystemer til forgasser-SOFC-systemer, da in-situ gasrens kan mindske/simplificere kravene betydeligt til gasrensningen nedenstrøms.

Den anden del af projektet udfører et komparativt studie af Viking-pilotanlægget, hvor den typiske luftblæste konstellation ændres til drift med O_2-CO_2. Det overordnede formål med studiet er at opnå en kvælstoffri produktgas og analysere de tilhørende driftsparametre, virkningsgrader og gaskvaliteter for driftstilstandene i forgasseren. Dette blev først undersøgt via termodynamisk modellering, hvor det ses at driftsparametrene er relativt tæt på dem for luft når der anvendes 21v% O_2-i-CO_2 som forgasningsmiddel. Næsten identiske parametre ses ved 30v% O_2. Pilotanlægget blev herefter modificeret og klargjort, og eksperimentelle tests med luft, 21v% O_2–i-CO_2 og 25v% O_2–i-CO_2 blev udført. Resultaterne validerede tendenserne fra modelleringsstudierne, da det ses at der ved 21v% O_2 opnås temperaturer for partiel oxidation og rist på 52-69^{o}C og 31-36^{o}C under dem for luft. Som forventet opnås der ved 25v% O_2 ligeledes

et fald, men med temperaturer tættere på luftblæst drift. Detaljeret gasanalyse viste at tjære- og svovlkoncentrationer på henholdsvis få mg/Nm^3 og <3ppm var lave på tværs af forgasningsmedier og at N_2-koncentrationen var reduceret til få procentpoint.

Tredje del af projektet analyserede muligheden for at udvikle stor-skala anlæg på >10MW_{th} baseret på To-trins forgasningskonceptet, og undersøgte om de kunne integreres i en polygeneration-kontekst henholdsvis. Fælles for de to studier var udviklingen af en bred vidensbase af bl.a. biomasseforgasnings-delprocesser, -state-of-the-art anlæg og tidligere To-trins opskaleringsstudier, som gennem kreative processer og termodynamisk modellering førte til studiernes resultater.
I første del af studierne blev potentialet for træbaserede, riste- og fluid bed-fyrede systemer analyseret. Blandt flere, blev to nye nævneværdige designtiltag implementeret i koncepterne: 1) anvendelse af en pyrolysereaktor med recirkulering af gas for indirekte anvendelse af tilgængelige varmekilder og for at undgå fortynding af gassen; 2) implementering af køling af partiel oxidations-gasserne med tørrerdamp, enten via direkte injektion eller via ejektordrevet gasrecirkulering. Dette for at muliggøre høje temperaturer og matche forgasningsreaktorens og brændslets tolerancer. Disse to tiltag førte til tre primære designkoncepter, der opnåede meget høje virkningsgrader og forventes at behøve minimal gasrensning nedenstrøms. De tre koncepter opnåede koldgasvirkningsgrader på 88-93%. Konceptet med to modstrømsreaktorer viste at være særligt interessant med dets høje virkningsgrad på 93% og øget støvtolerance i forhold til det nuværende To-trins-konfiguration. Selvom studiet stadig er i udviklingsfasen ses det, at koldgasvirkningsgraderne var 6-22%-point højere end de nuværende state-of-the-art-forgassere, samtidig med at der kun anvendes minimal eller meget simpel gasrensning i sammenligning.
I det andet studie blev der udført yderligere udviklingsarbejde, og brændsels- og driftsfleksibiliteten blev analyseret. Målet var at designe systemer, der kunne håndtere polygeneration med luft og 50v% O_2-H_2O. Tre anlægsdesign blev præsenteret: alle anvendte fluid bed-forgasningsreaktorer, men med enten en modstrøms-, langsom fluid bed- eller hurtig fluid bed-pyrolysekobling. For at udvide analysen, blev anvendelsen af træ og halm undersøgt for alle koncepter og driftstilstande. Resultaterne viser relativt stabile driftsbetingelser og koldgasvirkningsgrader som gennemsnitligt er 83-88%. Konceptet med en modstrømspyrolyse viste sig som det mest lovende koncept i forhold til simplicitet og koldgasvirkningsgrad. På linje med det første studie er det påvist, at disse koncepter på et teknisk grundlag er konkurrencedygtige med nuværende state-of-the-art-forgasningsanlæg i forhold til virkningsgrad, gasrensningsudstyr og brændselsfleksibilitet.

Polygeneration-koncepter er blevet foreslået af flere danske energiaktører og vil formentlig blive en del af det fremtidige energisystem frem mod 2030-2050 i 50-200MW_{th}-størrelsen. Baseret på dette projekt, konkluderes det at To-trins-polygeneration-konceptet er teknisk bæredygtigt fra et forgasningssynspunkt. Dette studie viser at forgasningssystemet kan modificeres til drift med både SOFC og iltblæsning uden særligt krævende tiltag i udstyr og drift. Derudover er nye, skalérbare To-trins koncepter blevet designet og modelleret, og har vist sjældent høje koldgasvirkningsgrader og driftsfleksibilitet. Derfor ses det foreslåede koncept som klargjort til udviklingen frem mod et bæredygtigt energisystem.

Structure of the thesis and publications

The thesis presents the full body of work of the Ph.D. project. It is structured around five scientific publications that I have main authored and provided the first manuscript draft for and reviewed – note that not all of the publications have completed peer-review yet. These articles are presented below and are denoted with roman numerals I to V and presented in Appendix 1-5. Besides the publications, a patent application related to Article IV has been filed and are given in Appendix 6. The thesis introduces the work in Chapter 1, provides the technical basis for the study in Chapter 2 and summarizes and draws conclusions and outlook in Chapter 8. Appendix 7-9 presents supplementary material for Chapter 2 and Appendix 10-13 presents model scripts related to Article III-V.

- Article I: Rasmus Ø. Gadsbøll, Jesper Thomsen, Christian Bang-Møller, Jesper Ahrenfeldt, Ulrik B. Henriksen: **Solid oxide fuel cells powered by biomass gasification for high-efficiency power generation**. Published in Energy (2017) vol. 131, p. 198-206. Presented in Appendix 1. I have been in charge of designing the experiments, operating the SOFC stack, data analysis and reporting, and to a lesser extend assisted with operating the gasifier.

- Article II: Rasmus Ø. Gadsbøll, Adrian Vivar Garcia, Jesper Ahrenfeldt, Ulrik B. Henriksen: **Solid oxide fuel cell stack coupled with an oxygen-blown TwoStage gasifier using minimal gas cleaning**. In review: Renewable Energy. Presented in Appendix 2. I have been in charge of designing the experiments, operating the SOFC stack, data analysis and reporting, and to a lesser extend assisted with operating the gasifier.

- Article III: Rasmus Ø. Gadsbøll, Zsuzsa Sárossy, Lars Jørgensen, Jesper Ahrenfeldt, Ulrik B. Henriksen: **Oxygen-blown operation of the TwoStage Viking gasifier**. Published in Energy (2018) vol. 158, p. 495-503. Presented in Appendix 3. I have been in charge of buying, testing/calibrating and commissioning the gasifier oxygen-modifications, designing the experiments, data analysis and reporting, and to a lesser extend assisted with operating the gasifier and analyze gas and tar samples.

- Article IV: Rasmus Ø. Gadsbøll, Lasse R. Clausen, Jesper Ahrenfeldt, Ulrik B. Henriksen: **Thermodynamic analysis of upscaled TwoStage gasifier concepts**. In review: Energy. Presented in Appendix 4. The publication phase was delayed +6 months due to the patenting process and therefore it has not been published yet. I have been in charge of planning and directing the joint work in the developmental phase, formulating the final products and calibrating and modeling the final systems along with optimization and reporting.

- Article V: Rasmus Ø. Gadsbøll, Lasse R. Clausen, Tobias Pape Thomsen, Jesper Ahrenfeldt, Ulrik B. Henriksen: **Flexible TwoStage biomass gasifier designs for polygeneration operation**. Published in Energy (2019) vol. 166, p. 939-950. Presented in Appendix 5. I have been in charge of carrying out and formulating the continued developmental work, calibrating and modeling the final systems along with optimization, parameter analysis and reporting.

- Danish and PCT (international) registered patent as one of three equally involved inventors (33% each). Rasmus Ø. Gadsbøll, Jesper Ahrenfeldt, Ulrik B. Henriksen: **A gasification unit, a method for producing a product gas and use of such a method**. No. PA2017 70775. The patent is related to a system in Article IV and is very shortly presented in Appendix 6. I have participated in the review/formulation of the patent text, presented it for patent office agents and pitched it on 2 occasions to potential industrial buyers.

Publications not included in the thesis

All publications stated have been peer-reviewed unless stated otherwise. The following ones are not included for evaluation:

- Co-authored article: Christoffer Ernst Lythcke-Jørgensen, Lasse Røngaard Clausen, Loui Algren, Anders Bavnhøj Hansen, Marie Münster, Rasmus Ø. Gadsbøll, Fredrik Haglind: **Optimization of a flexible multi-generation system based on wood chip gasification and methanol production.** Published in Applied Energy (2017) vol. 192, p. 337–359.

- Co-authored article: Joakim M. Johansen, Rasmus Ø. Gadsbøll, Jesper Thomsen, Peter A Jensen, Peter Glarborg, Paul Ek, Nikolai De Martini, Marco Mancini, Roman Weber, Reginald E. Mitchell: **Devolatilization kinetics of woody biomass at short residence times and high heating rates and peak temperatures.** Published in Applied Energy (2016), vol. 162, p. 245-256.

- Main-authored conference proceedings: Rasmus Ø. Gadsbøll, Jesper Thomsen, Christian Bang-Møller, Jesper Ahrenfeldt, Ulrik B. Henriksen: **Experimental analysis of a solid oxide fuel stack coupled with biomass gasification**. Published in Conference proceedings for the 23rd European Biomass Conference & Exhibition, 1-4 June 2015, Vienna, Austria. Peer-reviewed abstract only.

Relevant dissemination activities

Oral presentations:

- Experimental analysis of a solid oxide fuel cell stack coupled with biomass gasification. European Biomass Conference & Exhibition, Vienna, Austria, 2015
- Experimental analysis of a solid oxide fuel cell stack coupled with biomass gasification. DTU Sustain conference, Lyngby, Denmark, 2015
- Conditioning of gasification product gas. Lecture at the Danish Society of Engineers (IDA), Copenhagen, Denmark, 2017
- 4 presentations related to patenting process given to patenting agents and commercial industries, DTU, Denmark, 2017
- Design and analysis of upscaled TwoStage biomass gasifiers. European Biomass Conference & Exhibition, Copenhagen, Denmark, 2018
- 5 presentations at internal DTU seminars, DTU, 2015-2018

Teaching:

- 5 ECTS-point special course: SOFC operation on biomass producer gas, 2015. Student supervision along with Ulrik B. Henriksen and Jesper Ahrenfeldt.
- 5 ECTS-point special course: Two-stage gasification of sewage sludge, 2016. Student supervision along with Ulrik B. Henriksen and Jesper Ahrenfeldt.
- 5 ECTS-point special course: Fluidization of char for tar conversion, 2016. Student supervision and mentoring.
- 5 ECTS-point special course: SOFC operation with oxygen-blown TwoStage Viking gasifier, 2018. Student supervision and mentoring.

Table of contents

1. Introduction

1.1 Motivation and overall research project

In order to reduce the impact of climate change, the use of fossil fuels must be reduced to a minimum. However, because the global infrastructure, economy and demand are very dependent on these fuels it is vital to ensure cost-effective alternatives that can effectively phase out the intense use of carbon polluting sources relatively quickly. The most straight-forward path is to substitute renewable fuels into the current infrastructure, as this will keep costs relatively low from a system and operator perspective. In this framework, energy from biomass, *bioenergy*, is considered an essential component due to its flexibility on several platforms and global abundance [1][2][3].

Bioenergy is the most utilized renewable energy source, accounting for roughly 10% of the worlds energy demand and will likely be the main renewable source till 2050 [1][2][4]. The potential for vastly increasing the use of biomass is present, as the International Energy Agency estimate that the use bioenergy could be up to 24-42% of the worlds energy demand by 2050 [2][5]. The potential use of biomass is however a very complex subject that depends on a large number of factors. And while the global energy demand might be technically covered by biomass when not considering sustainability, the sustainable potential (defined as residues from forests and agriculture, surplus forestry, energy crops and increases in agricultural productivity) should be able to cover the biomass demand going forward - see Figure 1.1. However, because biomass is a limited resource and because the assumed sustainability of biomass *can* be fragile [2], conversion efficiency from biomass to product is of the essence.

Besides bioenergy, the energy demand will be met by a variety of energy sources. In recent years, the share of especially wind and solar power has increased rapidly, as they can provide cheap, renewable electricity [6]. In order to obtain the most economically sustainable transition towards a low-carbon energy system, these technologies are therefore essential going forward. Implementing large shares of fluctuating wind and solar power do however have significant drawbacks, as they can significantly stress the remaining energy system and will require storage solutions, careful grid management and flexible counterparts for balancing. Amongst several options, flexible, dispatchable power plants and power-to-gas applications via electrolysis are seen as very promising solutions. Especially biofuel and power-to-gas in combination is promising, as the addition of electrolytic hydrogen can roughly double the yield of biofuel for a given biomass input. In this framework, bioenergy can act as a key component, as it can balance out power fluctuations in carbon neutral power plants and link the power sector to other sectors including the transport sector by incorporating electrolytic hydrogen in biofuels.

Especially thermal *gasification* of biomass is an interesting technology as it provides a very efficient and flexible platform for processing a variety of biomasses and wastes into a *product gas* that can be used for a variety of products incl. heat, power and fuel production. It therefore has the potential to be a cornerstone of future energy systems, providing an efficient link between available resources, energy sectors (power, heat, transport) and energy demands. Gasification has been researched for many years and is partially commercialized, but further developments are still necessary in order for a large implementation to take place. These include increased conversion efficiencies from biomass-to-gas and gas-to-product, increased plant flexibility and development of large-scale plants.

Increased gas-to-product efficiency and product flexibility can be obtained by coupling gasification to *solid oxide cells* into a joint technological platform. These cells are very effective electrochemical reactors that can either operate as power producing fuel cells (SOFC) or power consuming electrolysis cells (SOEC). As the vocal point for this thesis, it will be shown that this combination of technologies can provide some of the most efficient power plants on the planet and enable storage of power in biofuels, whilst providing balancing of the power grid. Coupling gasification to solid oxide cells does however have some key challenges that this project seeks to address.

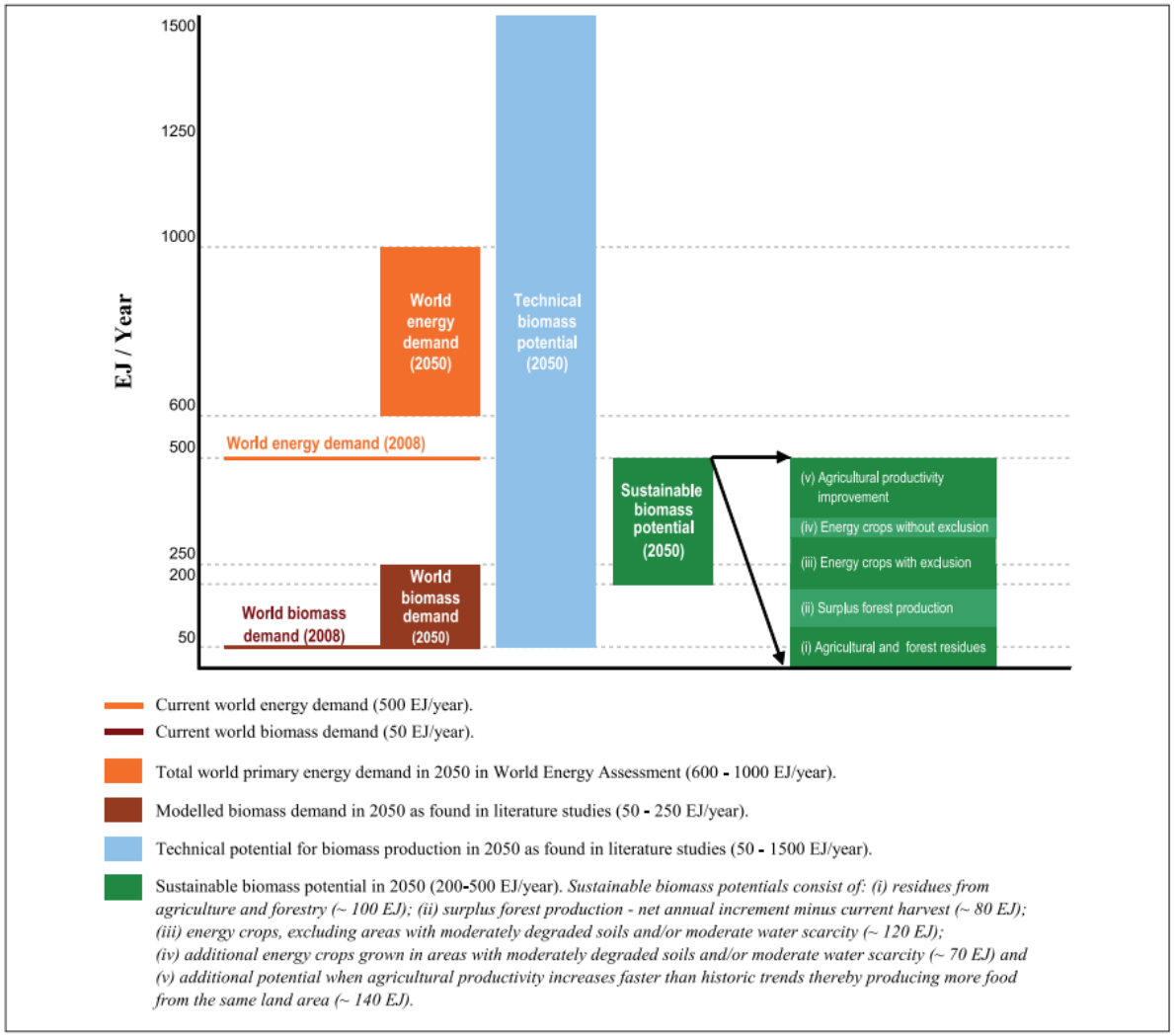

Figure 1.1 – The global outlook of primary energy demand and biomass potentials and demands [5].

1.2 Polygeneration

The research project is centered around a proposed *Polygeneration* concept that seeks to develop gasification technology. The concept is based on the coupling of solid oxide cells and thermal biomass gasification for which it is possible to design a system that continuously processes biomass into either power or power-assisted biofuel, depended on the immediate status of the grid. Hence the system can

either produce or consume power, which can be very beneficial for both the plant and the electricity grid operators' business cases, as power prices are projected to be increasingly fluctuating.

The Polygeneration system is conceptually shown in Figure 1.2. The gasification plant processes the solid biomass into product gas via partial combustion of the fuel with either air or oxygen. When producing power (high electricity prices), the gasifier uses air and the product gas is fed to a SOFC that electrochemically converts the chemical energy to electricity. When consuming power (low electricity prices), the solid oxide cell is used as a SOEC that converts steam and power into oxygen and hydrogen. The oxygen is used by the gasifier to produce a nitrogen-free product gas that is mixed with the produced hydrogen and added to a synthesis reactor that produces a biofuel and heat. Biofuels such as methane/synthetic natural gas (SNG) has shown to be compatible with this concept [7][8][9]. It is important to secure a nitrogen-free gas, as both gas separation and an over-dimensioned synthesis reactor are costly.

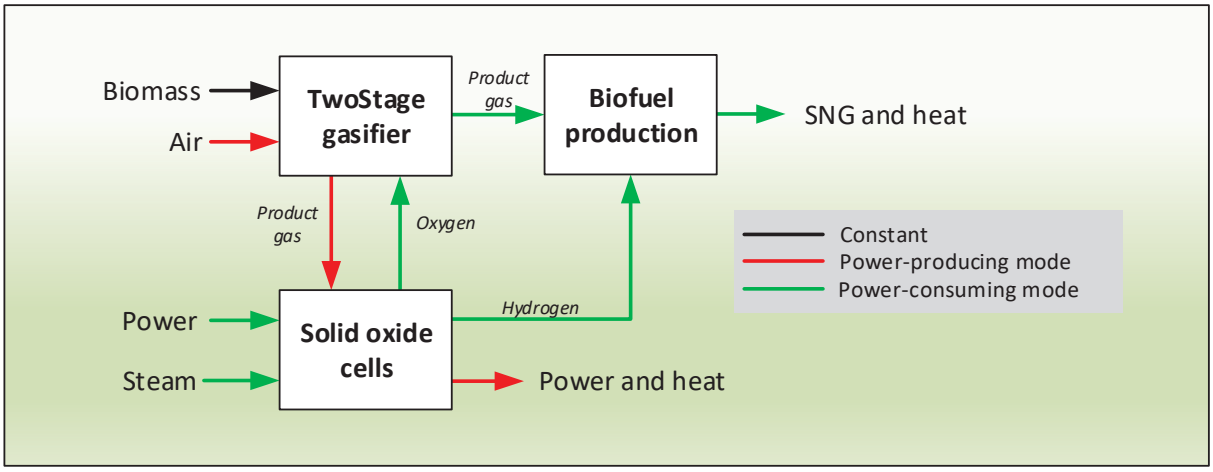

Figure 1.2 – Schematic presentation of the Polygeneration system

The Polygeneration concept has some distinct advantages:

- Biomass is a very flexible resource that can be harnessed for a variety of products. In contrast to wind and solar production, bioenergy is dispatchable.
- Gasification is very fuel flexible and can maximize the use of the biomass potential by potentially enabling resource such as straw and wastes as fuels
- Very high efficiency of gasifier and solid oxide cell: biomass-to-electric and biomass-and-power–to-biofuel efficiencies in the range of 43-63% and 69-70% (LHV) respectively has been found in modeling studies, which all are state-of-the-art results [7][9][10][11].
- Grid balancing made possible by either producing or consuming power, whilst enabling storage of power in biofuels that can be utilized in various sectors and stored long-term in the current infrastructure.

The proposed Polygeneration concept applies TwoStage gasification for biomass conversion, which has been developed for many years at the Technical University of Denmark (DTU). The gasifier is unique in its ability to convert wood chips into a very high quality product gas with negligible tar content at a very high cold gas efficiency, making it a state-of-the-art system. The gasifier is therefore very-well suited for the

Polygeneration concept that features SOFC and synthesis components that are very sensitive to a range of gas impurities. The TwoStage gasifier is however currently limited to primarily wood chips and small-scale of <10MW_{th}, which is namely due to the applied reactors/design of the system [12]. It is expected that the overall costs and/or efficiency can be improved at larger scales. Based on experiences with biomass power plants, reports has pointed to an optimum in the 50-200MW_{th}-range [2][5][13], which covers a range factors including process equipment costs, logistics, fuel accessibility and heat markets.

1.3 Objectives

Based on the previous chapters, this thesis' central research questions are:

- How can the TwoStage gasifier be effectively coupled to an SOFC and what is the potential?
- Can the existing TwoStage gasifier be effectively converted into utilizing oxygen instead of air?
- Can the TwoStage gasification concept be scaled up to medium-to-large capacities of >10MW_{th} and still maintain its low tar content and high efficiency? Can the fuel flexibility be increased to other fuels?
- Can the upscaled concepts be utilized effectively in a polygeneration context?

1.4 Methods

The project has been carried out in a series of steps based on the above objectives:

- Experimental coupling of the TwoStage Viking gasifier with a SOFC stack
    - Screen literature, analyze SOFC impuritity tolerances, assess gas cleaning options
    - SOFC tests with air-blown gasifier
    - SOFC tests with air and oxygen-blown gasifier including tests with minimal gas cleaning
- Experimental operation of an oxygen-blown TwoStage Viking gasifier
    - Screen literature, analyze oxygen-blown partial oxidation, assess gasifier performance
    - Modify the gasifier, procure, calibrate and connect equipment
    - Operating the gasifier with air and O_2-CO_2 mixtures
- Theoretical analysis' of the upscaling potential of the TwoStage gasifier
    - Screen literature, analyse subprocesses and design large-scale plant layouts
    - Thermodynamic modeling of plant layout and ongoing iterations to plant layouts
    - Evaluate select plants and compare to state-of-the-art alternatives
    - Design large-scale gasifier systems for Polygeneration operation

These steps has been completed in nearly chronological order as stated above. The methods have been formulated in five scientific articles and a patent, which constitutes the core of the work for this PhD project. Each of the articles represent individual pieces of research and hence contain their own literature survey, framing, analysis and results. This thesis then acts as a vehicle for connecting these publications by framing them in a larger context in the start (PhD formalia, project introduction and project technical background) and in the end (summarizing, analyzing and discussing across the studies and in a larger energy system perspective related to the initial project framing and subsequent developments).

1.5 Thesis outline

The thesis is structured in three main parts: Chapter 2 provides the technical background for the overall thesis; Chapter 3-7 presents the core of the research carried out in the form of scientific articles; Chapter 8

summarizes the individual research and achievements, whilst also constructing general conclusions across the studies and the current knowledge base on polygeneration.

The written content of the introduction (Chapter 1), technical basis of the study (Chapter 2) and concluding remarks (Chapter 8) is mainly original, but some subsections and phrases from the listed publications (that I have authored) are re-used in these chapters.

2. Technical basis for the study

This chapter will present the background of the overall project, including biomass gasification, TwoStage gasification and solid oxide fuel cell technologies. Specific literature reviews on the performed studies are included in the publications in the following chapters.

2.1 Biomass gasification

Thermal gasification is the high-temperature thermochemical conversion of carbonaceous matter into product gas. The gasification platform enables the use of many different bioressources including wood, agricultural residues and wastes that can be very efficiently converted to gas. State-of-the-art gasifier plants typically reach cold gas efficiencies of 75-93% [14]. All of the fuels can be processed into either heat, power, chemicals, fertilizers and fuels via several applications. Hence, this platform offers a unique opportunity to make a very cost-effective transition from fossil fuels via very high fuel flexibility, conversion efficiency and product flexibility.

The conversion from solid biomass to product gas is carried out through a series of steps: drying, pyrolysis, gasification and combustion – see Figure 2.1. These reaction steps can either be carried out in a single reactor or multiple reactors. The process produces a variety of species including permanent gases, condensable tars and solids, all of which can be used for several purposes. A large span of fuels can be gasified, including solid and liquid biomasses and fossil fuels. Drying will not be covered here.

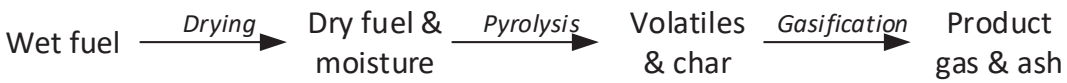

Figure 2.1 – Conversion steps of fuel to product gas

2.1.1 Pyrolysis

Pyrolysis is the decomposition of the fuels initial macrostructure, breaking the main components of the fuel into smaller organic molecules (C-1 to C-6 hydrocarbons), larger organic molecules named *tars* (covered in section 2.1.3), and light permanent gases (mainly H_2, CO, CO_2), whilst producing solid char consisting of primarily fixed carbon and ashes. The process consists of a very large number of reactions that, without reacting with an oxidizing atmosphere, summarises into a practically balanced global reaction that is either slightly endothermic/exothermic. Devolatilization does however have a heating demand of 4-7% of the input fuel (LHV) as the fuel needs to be heated up [15].

Pyrolysis can be divided into three sub-processes at separate temperature levels [16]:

- At temperature of 150-300^{o}C, the macromolecules of biomass - hemi-cellulose, cellulose and lignin - start to activate and dehydrate. This part is characterized by the initial breakdown of the structure, which causes the fuel loses its fibrous structure and makes it brittle.
- At temperatures >200^{o}C the main pyrolysis occurs, breaking the down the macrostructure of the biomass. The process releases light gases and major condensable organic molecules named tars. The main release of tars and total volatiles are carried out till ≈500^{o}C and ≈600^{o}C respectively [17].
- At temperatures between 300^{o}C and >600^{o}C several secondary reactions occur between the products of the primary pyrolysis. Released volatiles are subject to cracking or reforming, yielding

lighter gases and secondary char. The reactions are caused by decomposition by temperature, gas-phase reforming reactions and chemical reactions with the primary char [18].

2.1.2 Char gasification

The gasification reactions convert the solid carbon in the produced char into a gaseous fuel by a series of exo- and endothermic gas-solid reactions. The solid dry char consists of approximately 50-75wt% carbon, 10-20 wt% oxygen, 15-40 wt% ash and traces of hydrogen [19] - the reactions are here stated traditionally as a reaction with pure carbon. The reactions and their reaction enthalpies are shown in Equation 2.1 to Equation 2.5. The combustion reactions are the typically main driver of the gasification and pyrolysis, as they provide the necessary heat for the pyrolysis and the endothermic char reactions with steam and CO_2 – these reactions might be considered as pure combustion reactions and not gasification reactions. Hence gasifiers are typically air-blown to drive the endothermic reactions, but can utilize O_2, H_2O, CO_2 or a mixture of these gases, which can be beneficial if e.g. N_2-dilution is unwanted.
Temperatures of >700°C are usually required in order to process the char at a satisfying rate [16][19]. It should be noted the reaction rates of the solid-gas reactions sorted are: O_2 >>> H_2O > CO_2 >> H_2, with O_2 achieving an order of 3-5 times higher rates then H_2O and 6-7 times that of CO_2 [16]. The H_2 reaction rate is by far the slowest and is typically negligible.

$$C(s) + \frac{1}{2}O_2 \rightarrow CO + 111\frac{kJ}{mol}$$
Equation 2.1

$$C(s) + O_2 \rightarrow CO_2 + 394\frac{kJ}{mol}$$
Equation 2.2

$$C(s) + H_2O \rightarrow CO + H_2 - 131\frac{kJ}{mol}$$
Equation 2.3

$$C(s) + CO_2 \rightarrow 2CO - 172\frac{kJ}{mol}$$
Equation 2.4

$$C(s) + 2H_2 \rightarrow CH_4 - 75\frac{kJ}{mol}$$
Equation 2.5

During gasification (and pyrolysis) there is a host of freeboard gas-gas reactions including decomposition/reforming of tar molecules, reforming of lighter gases and partial combustion. Of all of these reactions, the far most important is the water-gas shift reaction (see Equation 2.6), which is typically used to predict the product gas composition in gasifier models.

$$CO + H_2O \rightarrow CO_2 + H_2 + 41\frac{kJ}{mol}$$
Equation 2.6

2.1.3 Tars and conversion measures

Tars are organic fragments that originate from the original fuel structure that are released during pyrolysis. They are a large and very complex group of compounds, ranging from simple alkane oxygenated alcohols to large polyaromatic hydrocarbons (PAH), but are typically consisting of a variety of aromatic compounds such as phenols and naphthalenes [20]. While there has been much debate on the classification of tars, the

IEA Gasification Group have officially defined tars as "*hydrocarbons with molecular weights higher than benzene*" [21].

Tar compounds are a vocal point for gasifier and pyrolysis designs, as they can be very troublesome for the plant operation if not handled correctly. This can manifests itself in limited plant availability, expensive and complex gas cleaning equipment and/or a loss of chemical energy combined with waste water management. Hence tars are considered one of the main challenges for broad commercialization of the technology. Typical areas of concern for tars are:

- Tars can easily condense and clog equipment as they typically outcondense between 200-400^{o}C (but the dew point is of course dependent on the tar concentration in the gas). Even smaller concentrations can hence interrupt operation as pipes and valves are clogged over time.
- The tar content of the product gas can contain a significant part of the chemical energy released from the fuel. The high energy content of tars makes them vital to the energy efficiency - whether they are used as a fuel (bio-oil), removed or converted to gas in order to obtain better gas quality.
- Catalysts can be fouled, as the breakdown of tars is associated with high soot formation that can block active sites of the catalyst and increase the pressure drop. Continuous catalyst regeneration is therefore necessary to maintain catalyst activity.

In order to avoid the problems with tars, the content should be lowered to nearly negligible levels for most applications including engines and synthesis. The tar content can be reduced and converted via a variety of measures. Typically, a combination of measures is needed in order to reduce the tar content to negligible levels and secure high gas quality. The main tar reducing measures include physical removal, thermal/oxidative conversion and catalytic/chemical conversion. Physical removal is a well-proven method for outcondensing tars and can be carried out via filters and scrubber using water/oils. Scrubbers are widely applied as the final step of a gas cleaning train in order to remove remaining tars, but waste water management can difficult if tars are not separated and fed into the system again. Tars should however be converted to permanent gases if very high cold gas efficiencies are to be had and hence physical removal techniques are not discussed further here. The reader is referred to e.g. [20][22] for details on physical removal of tars.

Partial oxidation

Thermal and oxidative treatment of tars via partial oxidation (POX) of gas is of significant interest as it is very effective, simple and can be integrated in the process with no or little loss energy loss. The main tar conversion step in gasifiers is the typically applied temperatures of ≈800-900^{o}C and added oxygen, that significantly reduces the tar content and also changes the speciation towards larger (PAH) tar molecules via thermal treatment and oxidation [23][24]. Both measures convert tars by breaking the chemical structure of the tar molecules and converting them into light permanent gases (H_2, CO, CO_2, CH_4), and soot, whilst changing the remaining tar species towards more deoxygenated and cyclic/aromatic species [25]. Higher temperatures and oxidant will increase this conversion.

While tars can be converted inside a single reactor via these ‘primary measures’, the gasifier can be designed to feature a separate reactor that can partially oxidize a gas flow. This reactor can be placed either between pyrolysis and gasification or downstream the gasifier. In staged gasifier systems with separate pyrolysis and gasification, POX is a very powerful tool, as the tarry pyrolysis gas can be treated

without a loss of system efficiency[1]. This can happen at very high temperatures of ≥1100°C that often can convert 95-99% in such a step [26]. Even at lower temperatures of 900°C, POX can be very efficient if the design and gas-gas contact is combined with an adequate air flow: Iversen et al. [27] suggested the POX to be operated at ≥900°C and a stoichiometric air ratio of ≥0.4 for high reductions of a variety of tars including phenol- and naphthalene-like species. Table 2.1 presents an overview of measurements from previous tests - the wide spread in data should be noted, indicating that the various operating/burner conditions are quite diverse and important. It can be seen that at 1200-1300°C very little tar remains in the gas.

Temperature [°C]	Stoichiometric air ratio	Tar content [mg/Nm^3,dry][a]	Note	Reference
900	0.5	115	POX	[26]
900	0.34	960	POX	[28]
900	-	2844	POX	[29]
1050	0.4	≈846[b]	POX	[24]
1100	0.34	1000	POX	[30]
1100	-	1150	POX	[31]
1100	0.5	1200	POX	[32]
1100-1200	-	1220	POX	[33]
1200-1300	-	≈100	POX	[31]
1000	0	5000	Thermal treatment	[26]
1100	0	8000	Thermal treatment	[30]
1200	0	385	Thermal treatment	[34]
1250	0	50	Thermal treatment	[34]
1300	0	10	Thermal treatment	[34]

Table 2.1 – Overview of partial oxidation and thermal treatment studies. [a]Based on 2.6Nm^3/kg gas production from biomass, as for the TwoStage gasifier [32]**. [b]Estimate.**

Chemical and catalytic conversion

Converting tars by chemical and catalytic means is very appealing, as they can be very effective, operate at reduced temperatures and have excellent energy efficiency, as they similar to POX convert tars into permanent gases and soot. Tars can be effectively converted down to 750-850°C dependent on conditions, which matches very well with general gasification and raw product gas temperatures and is a major attraction. And while there have been much research in this field, this study will only deal with the most applied and relevant ones: char, dolomite, olivine and nickel catalysts – the reader is referred to e.g. [20][22][35] for more details on other options. These four options for this study are described in detail in Appendix 7.

The discussed catalysts can be applied in either fixed or fluid beds. Fixed beds typically offer better gas-solid contact, as especially the bubble formation in fluid beds can prevent tar-catalyst mixing and residence times typically are lower, which result in lower tar conversion. Fluid bed catalysts are however attractive for a number of reasons (e.g. fuel flexibility and scaling) and this project has therefore included research on

[1] While combustion reduces the chemical energy content, the applied POX conditions are not enhanced in any way for the sake of tar conversion, but is the result of the design parameters as e.g. providing enough thermal energy for the char gasification. If the released thermal energy is not utilized for char gasification or elsewhere in the fuel conversion, the cold gas efficiency will of course drop.

means to improve the gas-solid contact in fluid beds. A total of 6 reactor configurations are presented in Appendix 8, based on studies of hydrodynamics and literature on tar conversion.

Char is an attractive way to reduce tars as char is produced inside the gasifier and is very active at high temperatures. Char also has the ability to recover thermal energy from the partial oxidation by endothermic char gasification reactions and thus functions as a chemical quench. Besides that, char beds are also efficiently coupled with partial oxidation because char can convert heavier tars e.g. PAH that form at the high POX temperatures [32][36][37]. Tar reduction over char is a proven technology for fixed beds with very high conversions - converting 95-99% of the tars inside the gasifier at 850^{o}C and 0.3-1.2 s of residence time [32][36].
In fluid beds, char is relatively unproven, as it exhibits low mechanical strength and suffers from a high attrition rate. But tests with fluid beds by El-Rub et al. [36] with char as the only bed material have shown high reductions at 850-900^{o}C, with gasifier tar concentrations reduced by >90% compared to sand. As a downstream fluid bed reactor, tar reduction only reached 70-80% when naphthalene was employed as a synthetic tar at 900^{o}C [36].

Catalytic conversion of tars via dolomite, olivine and nickel is widely investigated and applied. Dolomite and olivine has the distinct advantage of being cheap and abundant natural minerals that can be very effective if the mineral composition, pretreatment and applied conditions are chosen carefully. While they are closely related, dolomite offers the superior performance with regards to tar conversion, which can be readily compared with char and can achieve 99% tar conversion. The main advantage for olivine is that it possesses a mechanical strength similar to sand, which makes it attrition resistant and very suitable for fluid bed operation, in contrast to the brittle dolomite [38]. Hence the slightly lower tar conversion, of typically 10-20% lower than dolomite [23][39], is tolerated. Olivine has been found to be significant less active when converting larger tars such as naphthalene – 50-80% [40] and 55% [36] conversions has been found at 900^{o}C at similar residence times of 0.3 seconds. Nickel catalyst are widely applied in commercial systems, as they are very effective in converting tars and can do so at ≥100^{o}C lower temperatures than the mentioned natural minerals. Typically >98% of tars can be removed from ≥730^{o}C [41][42][43]. While very effective, nickel catalysts are quickly deactivated if inorganics and soot formation is not controlled. With some uncertainty due to different operating and catalyst conditions, the tolerance to sulphur is in the range of of 100ppm sulphur [44][45][46] and tars around 2g/Nm^{3} [41][47] (see Appendix 7 for further details).

A comparison of the four studied tar converters is given in Table 2.2.

	Char	Dolomite	Olivine	Nickel
Relative mechanical strength	Low	Low	High	Depends
Preferred reactor configuration	Fixed bed	Fixed bed	Fluid bed	Fixed bed
Tar selectivity	All/heavy	All	Light	All
Max conversion	99%	99%	55-97%	99%
Max conversion, lower temperature	850°C	850°C	820-900°C	750°C
Relative price	Free/Low	Low	Low	High
Main disadvantage	Low strength	Low strength	Low conversion of heavy tars	Deactivation from soot and inorganics

Table 2.2 – Comparison of tar conversion compounds with approximate parameters.

2.1.4 Gasifier types

Gasifiers come in a lot of different shapes and sizes dependent on the fuel, scale, application and operator. Different reactors each represent their unique way of processing the fuel from solid to gas and ashes with regards to e.g. temperature, particle size, residence time, efficiency. Characterized by their bed configuration, the main gasifier types are fixed beds, fluid beds and entrained flow. A detailed description of each gasifier type can be found in Appendix 9.

Fixed beds are mainly characterized by being relatively simple and efficient and therefore the most applied gasifier type. They are however limited by fuel flexibility and scalability, which primarily limit them to wood chip/pellet operation and up to single MW_{th}-capacity for downdraft and tens of MW_{th} for updraft units [48]. Fluid beds are somewhat more complex and less efficient than fixed beds, but provide excellent fuel flexibility and good scalability via excellent heat transfer and mixing. The main challenge for fluid beds is tars, which are only partially converted in the system and thus several steps of gas cleaning are typically required. Of all of the types, the entrained flow gasifier is potentially the most market-ready technology, as it via a single reactor step can convert a variety of fuels into a high quality gas and melted slag. Easy pressurisation via slurry feeding is also very attractive for synthesis and gas turbine applications. The main drawback is the lower efficiency and loss of potentially valuable inorganics in the slag. The different types are compared in Table 2.3. Equation 2.7 defines the cold gas efficiency.

	Updraft	Downdraft	Bubbling fluid bed	Circulating fluid bed	Entrained flow
Fines tolerance	Moderate	Low	High	Very high	Very high
Coarse tolerance	Very good	Very good	Moderate	Low-moderate	None
Fuel flexibility	Wood, some pelletized fuels[a]	Wood, firmly pelletized fuels[a]	Wood, straw, wastes etc.	Wood, straw, wastes etc.	All grindable
Oxygen demand	Low	Low-moderate	Moderate	Moderate	High
Gas-solid contact	High	High	Moderate	Moderate	Low/none
Tar content of product gas	High	Low	Moderate	Moderate	Trace
Typical maximum temperature	1000°C	1000°C	750-900°C	750-900°C	≥1300°C
Fuel conversion	High	High	Moderate	Moderate	High
Cold gas efficiency[b] (incl. tars)	90%	85%	80-85%	80-85%	72-80%[c]
Scalability limit	Tens of MW_{th}	Single MW_{th}	Hundreds of MW_{th}	Hundreds of MW_{th}	Single GW_{th}

Table 2.3 – Overview of gasifier types and approximate parameters. [a]Fuels should be firmly pelletized to avoid excessive fines and ash softening temperatures should match the specific system. [b]State-of-the-art efficiencies for single reactors incl tars, are defined via Equation 2.7 where PG denotes product gas. [c]The reactor typically reaches 80%, but will likely require torrefaction for pulverizing the fuel that requires up to 10% of the heating value of the fuel.

$$\eta_{cg} = \frac{\dot{m}_{PG} \cdot LHV_{PG}}{\dot{m}_{fuel} \cdot LHV_{fuel}}$$

Equation 2.7

2.1.5 TwoStage gasification

At the Technical University of Denmark, the Biomass Gasification Group has been developing the TwoStage biomass gasification process for many years. It is a gasification system designed with separate pyrolysis and gasification and a partial oxidation in between the two. The gasifier can process wet wood chips into a syngas with a very low tar content. The process has been heavily investigated and documented at smaller scales up to ≈1.5MW_{th} and has displayed its superiority to other systems within several aspects [49]. Especially the internal tar conversion, that greatly simplifies the gas cleaning, and the very high cold gas efficiency are key parameters for the system.

The current TwoStage design uses an indirectly heated screw conveyer for pyrolysis, in which hot product gas or engine exhaust heats up the wood fuel to 600°C through a metal jacket. Following the pyrolysis, the gases are exposed to a POX with air that increases the temperature to >1100°C and reduces the tar concentration to 1.0-1.5g/nm^3 [31][32][33]. The hot products are then led through a downdraft char bed resting on a grate in order to gasify the char, where only 0.1mg/Nm^3 naphthalene has been detected

before gas cleaning (filter) [49][50]. See Figure 2.2 for an overview of the TwoStage 'Viking' gasifier plant. By converting tars internally and utilizing the sensible heat from either product gas or engine exhaust (as in Figure 2.2) to dry and pyrolyze the fuel, the cold gas efficiency becomes very high - 93.2%(wet basis) has been measured [49]. The sulphur content of the product gas has been previously measured to 0.93ppm of COS and 0.5-1.0ppm of H_2S [11][51], and 3.7ppm of COS [10] respectively. A typical product gas composition is given in Table 2.4.

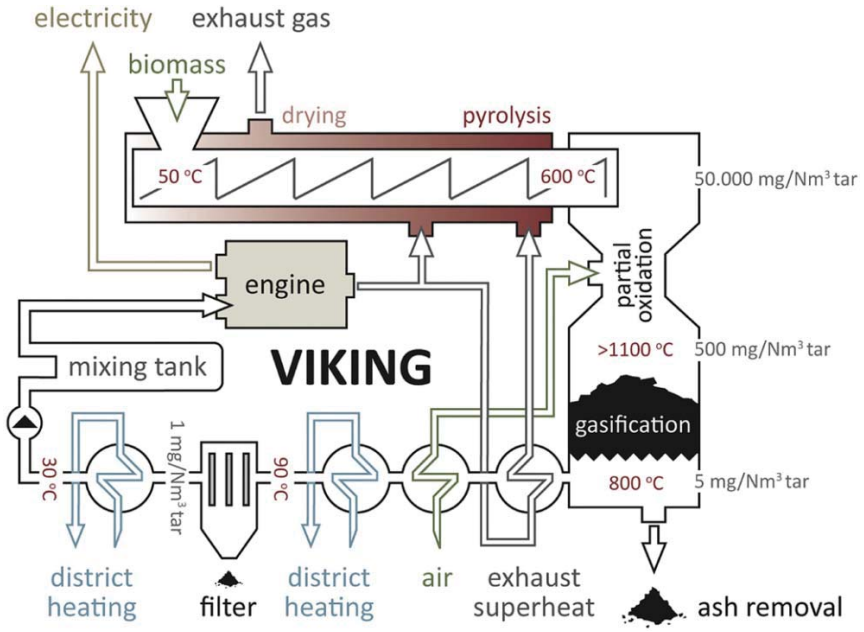

Figure 2.2 - Flow diagram of the TwoStage Viking gasifier at the Technical University of Denmark with approximate operational parameters [52]

Gas component	Dry gas composition
CO [vol%]	19.6
CO_2 [vol%]	15.4
H_2 [vol%]	30.5
CH_4 [vol%]	1.2
N_2 [vol%]	33.3
H_2S+COS	<4ppm
LHV_{gas} [MJ/kg]	6.2

Table 2.4 – Typical product gas composition from the TwoStage Viking gasifier [10][11][49][51]

2.2 Solid oxide fuel cells

SOFC technology is an interesting option for near-future high-efficient power generation systems. The high efficiency, typically of 45-60% (and higher with system integration) [53][54] is the main driver compared to other power producing technologies such as engines and turbines. SOFC is a developing technology platform, but it is typically operated at high temperatures in the range of 650-1000°C. The high

temperatures are both the source of their strengths and weaknesses: no need for expensive catalysts, increased kinetics/performance and fuel flexibility, weighed against decreased lifetime and complex manufacturing. As seen in
Table 2.5, the SOFC possesses significant advantages to other fuel cell types, which makes them a vocal point for research.

Advantages	Disadvantages
Very high efficiency	Low lifetime
High quality waste heat for system integration	Complex ceramic materials
No expensive catalysts	High manufacturing costs
High fuel flexibility: can use CO and hydrocarbons	More complex plant design at high temperatures
More tolerant to gas impurities	

Table 2.5 – SOFC compared to other fuel cell types

Working principle

A fuel cell is an electrochemical reactor that can convert the chemical energy captured in gaseous fuels directly into electricity. The fuel cell utilizes separate flows of fuel and oxidizer and an ion-conducting membrane to produce electricity directly via electrochemical reactions. Because of this characteristic it is not subject to the limits of the Carnot cycle as traditional heat engines, but instead traditionally[2] to the limits of the relationship between the change in Gibbs free energy and the change in enthalpy of formation (the calorific value). While this does not make the fuel cell more efficient than heat engines in all cases, this mechanism enables the fuel cell to operate at very high thermodynamic efficiency that typically outperforms that of heat engines.

All fuel cells consist of three main parts: two porous, electric-conducting electrodes connected with a wire in a circuit and an ion-conducting, electric-insulating and gas tight electrolyte. The cell features a negatively charged (anode) and positively charged (cathode) electrode and is where the fuel and oxidizer (air or oxygen) are added respectively. A general working principle of a SOFC is shown in Figure 2.3.

[2] The fuel cell efficiency can be defined in a number of ways. The reader is referred to e.g. [55] for additional discussion and details.

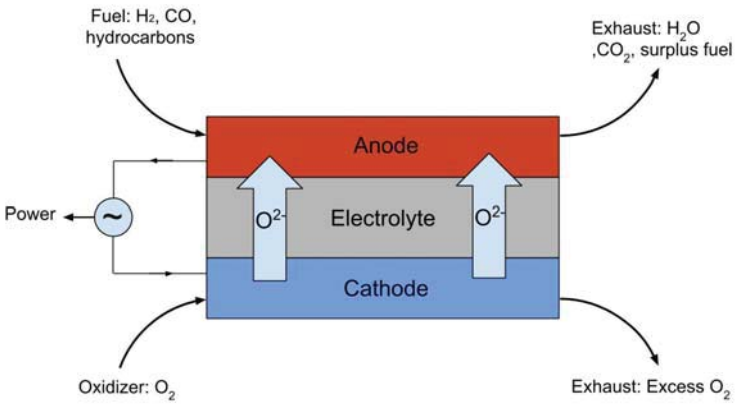

Figure 2.3 – Working principle of the SOFC

The SOFC consists only of metallic and ceramic materials that can withstand high the temperatures. In the SOFC air is added to the cathode, where the oxygen-molecules are split and reduced by absorbing electrons delivered from the electric circuit (Equation 2.8).

$$O_2 + 4e^- \rightarrow 2O^{--}$$

Equation 2.8

The oxygen-ions can then move across the electrolyte to the anode. The electrolyte is typically an alloy of yttrium stabilized zirconia that at high temperatures can conduct oxygen-ions at relatively high levels. At the anode, the gaseous fuel (H_2 and CO) is added and reacts with the oxygen-ions (Equation 2.9 and Equation 2.10). The overall reversible reaction in the fuel cell is then given in Equation 2.11, with *Energy* being the combined change in Gibbs free energy of formation of the reactions and waste heat. The change in Gibbs free energy is released as electricity, with the flow of electrons in the system. The irreversibilities in the cell cause a production of heat that is released from the system, either through external heat loss or mainly through internal heat exchange with the air feed.

$$H_2 + O^{--} \rightarrow H_2O + 2e^-$$

Equation 2.9

$$CO + O^{--} \rightarrow CO_2 + 2e^-$$

Equation 2.10

$$H_2 + CO + O_2 \rightarrow H_2O + CO_2 + Energy$$

Equation 2.11

In order to maintain stable and uniform operation of the fuel cell and avoid anode oxidation, fuel is added in slightly excessive amounts in order to avoid too large fuel gradients through the cell. The amount of fuel used, or *fuel utilization* factor, must be carefully optimized in order to ensure high performance [55]. The unprocessed fuel from the anode exhaust should to be processed by e.g. recirculation or by a downstream application.

The high-temperature fuel cell can besides H_2 and CO use CH_4 and other hydrocarbons as fuel via internal reforming. Due to the high temperatures the methane is reformed internally in the cell (usually by adding a

nickel catalysts and steam [56]) into H_2 and CO by steam reforming - shown in Equation 2.12. Caution must be taken when operating on hydrocarbons as soot, C(s), can form deposits on the anode – shown in Equation 2.13. In order to prevent soot build-up, a sufficient gas composition that can either prevent carbon deposition via the equilibrium or gasify the carbon with either steam and/or CO_2 (Equation 2.3 and Equation 2.4) should be designed. Steam is typically added to high-temperature systems using H_2-CO mixtures, as carbon deposits can be formed through the inverse Boudouard-reaction (Equation 2.14).

$$CH_4 + H_2O \rightarrow 3H_2 + CO$$
Equation 2.12

$$CH_4 \rightarrow 2H_2 + C(s)$$
Equation 2.13

$$2CO \rightarrow CO_2 + C(s)$$
Equation 2.14

2.3 Solid oxide electrolysis cells and reversibility

This project is limited within the polygeneration framework to full-chain electricity production with SOFC and oxygen-utilization in the gasifier system. Hence, the fuel cell configuration is the primary focus of this project, but a very short presentation of its reverse mode - the solid oxide electrolyzer cell (SOEC) configuration - is given here. For detailed SOEC technology and reversible SOFC/SOEC operation information the reader is referred to [57][58][59][60] and [60][61][62] respectively.

The SOFC/SOEC is very similar with regards to the overall construction, properties, operation and temperatures. Opposite the SOFC, the SOEC converts steam and/or CO_2 into O_2 and H_2/CO. The working principle is shown schematically in Figure 2.4, where H_2O and/or CO_2 is split at the cathode and oxygen-ions transported to the anode through the electrolyte membrane. Electricity-to-fuel efficiencies are projected to be as high as 77-90% (LHV) going forward, which is significantly higher than other electrolyzer types [57].

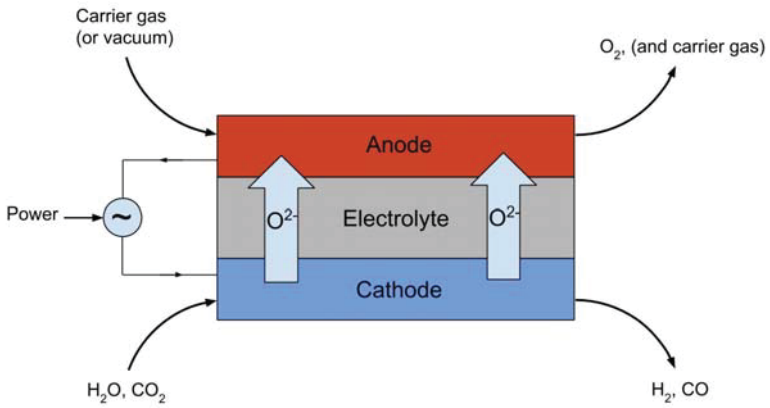

Figure 2.4 – Working principle of the SOEC

The key difference compared to SOFC is that the process is generally endothermic, consuming both steam/CO_2, electricity and heat[3], and processing it into fuel and oxygen. Initial reactions of H_2O and/or CO_2 at cathode with the addition of electrical energy are given Equation 2.15 and Equation 2.16.

$$H_2O + Energy \rightarrow H_2 + O^{--}$$

Equation 2.15

$$CO_2 + Energy \rightarrow CO + O^{--}$$

Equation 2.16

Oxygen-ions then form O_2 at the anode and release electrons back to the circuit. The oxygen can be then be extracted via a carrier gas or sucked from a vacuum. The global reaction is shown in Equation 2.17.

$$H_2O + CO_2 + Energy \rightarrow CO + H_2 + O_2$$

Equation 2.17

[3] Assuming that the system operates below the thermoneutral voltage. At the thermoneutral voltage the system is in balance and no heat addition is needed, and electric conversion efficiencies are very high [59]. Above this voltage the operation shifts towards exothermic operation.

3. Article I: Solid oxide fuel cells powered by biomass gasification for high efficiency power generation

Appendix 1 present the published article.

4. Article II: Solid oxide fuel cell stack coupled with an oxygen-blown TwoStage gasifier using minimal gas cleaning

Appendix 2 present the article currently in review

5. Article III: Oxygen-blown operation of the TwoStage Viking gasifier

Appendix 3 present the published article. Appendix 10 presents the DNA model script.

6. Article IV: Thermodynamic analysis of upscaled TwoStage gasifier concepts

Appendix 4 present the article currently in review. Appendix 11 presents the DNA model scripts.

7. Article V: Flexible TwoStage biomass gasifier designs for polygeneration operation

Appendix 5 presents the published article. Appendix 12 presents the DNA model script. Appendix 13 presents the results in the form of a data file that accompanies the publication.

8. Concluding remarks

8.1 Summary of findings and conclusions

This thesis' central research questions were focused on:

- How can the TwoStage gasifier be effectively coupled to a solid oxide fuel cell (SOFC) and what is the potential?
- Can the existing TwoStage gasifier be effectively converted into utilize oxygen instead of air?
- Can the TwoStage gasification concept be scaled up to large capacities of >10MW_{th} and still maintain its low tar content and high efficiency? Can the fuel flexibility be increased to other fuels?
- Can the upscaled concepts be utilized effectively in a polygeneration context?

These led to:

- Two experimental campaigns where the TwoStage Viking gasifier was coupled to a SOFC stack where the technical feasibility with regards to peak and part load performance, and level of gas cleaning was assessed with two different product gases.
- Experimental work was also carried out as the gasifier was modified for O_2-CO_2 operation. Here, a comparative study with air was carried out to evaluate the performance of the system with special focus on the system operating temperatures and the concentrations of tar and sulphur in the product gas.
- Finally, large-scale system design were developed on the basis of an analytical and creative process, which resulted in a larger assessment of the developmental potential for the TwoStage gasifier system in fixed and fluid bed versions. Analysis of the performance on a 1st and 2nd law basis, and flexibility with regards to wood/straw and air/O_2-H_2O were carried out in two modelling studies respectively.

The basis for the study was built through a vast literature study, as the topics ranged from handling of oxygen, to fuel cell mechanisms, to gas cleaning, to detailed reactor conditions, to plant design. Hence a key of this project has been the development of a broadspectred and deep academic foundation. This were especially true for the plant design, as the optimal development of novel gasifier systems required insights into, not only state-of-the-art technology and reactor conditions, but also an assessment of future energy scenarios (EU, DK), plant economics, low-grade fuel potential and conversion, and evaluation of material stresses and lifetimes of the subsystems. Hence both physical and strategic constraints were evaluated.

TwoStage gasification-SOFC coupling

The potential of coupling the TwoStage gasifier to a SOFC had previously been studied with expectations that the system could significantly outperform existing alternatives on the small-scale market. This project hence coupled these state-of-the-art subsystems and in Article I presented 145 hour proof-of-concept demonstrations with no significant/visual degradation of the SOFC at nominal conditions. It was shown that the peak performance was in line with the modelled results: showing electric efficiencies (gas-to-power) up to 46% for the SOFC, which was found to be the highest reported results in the literature, and an estimated ≈40% biomass-to-power efficiency for the overall system. Hence a new state-of-the-art bar has been set. The system also showed excellent part-load performance of the stack down to 55% flow without loss of electric efficiency, which is an increasingly important characteristic for power plants. Net electric

efficiencies have previously reached 25% with gas engines coupled to the Viking plant, with expectations of an achievable 35% with a modern engine [14].

The technical feasibility was further investigated in Article II, where the impact of applying minimal or no gas cleaning between the systems was studied along with operational characteristics at various temperatures and product gas compositions. Initially, the gas cleaning train from Article I was simplified to a carbon filter and desulphurizer and tests were performed with an air- and O_2-CO_2-blown product gas. A total of 4 tests were performed. Test 1 and 2 tested SOFC operating temperatures of 700^{o}C and 800^{o}C with an air-blown product gas, and found power and efficiency increases of 8-11% and 4%-points respectively at 800^{o}C. The gas-to-power SOFC efficiency was found to 40% at a fuel utilization of 69%. Test 3 and 4 implemented an O_2-CO_2-blown product gas (as produced in the study in Article III) and the high-CO containing gas is found in Test 3 to effect the performance negatively due to lower CO diffusion rate. In Test 4, the carbon filter and desulphurizer were bypassed during open-circuit conditions and no short-term changes in voltage was seen with 1.5-2.8ppm sulphur in the feed gas. A polarization curve mapping showed no significance on the performance when bypassing the gas cleaning. This indicates that the gasifier design can be a key feature when constructing gas cleaning trains for gasifier-SOFC systems, as in-situ gas cleaning can reduce the downstream cleaning significantly.

Oxygen-blown operation of the TwoStage gasifier

With the motive of producing a N_2-free product gas, the 80kW$_{th}$ Viking gasifier was modified from using air to an O_2-CO_2 blend as gasification medium. Article III presented the process of carrying out the conversion and presented the experimental campaign. Initial planning was carried out via a thermodynamic modelling study, which investigated the effects of applying the new media. The study found that operating conditions with 21v% O_2-in-CO_2 were expected to be in the range of air-blown values, but with lower performance. As the concentration reached 30v% O_2, the system is expected to reach nearly identical operation conditions to air including partial oxidation (POX) temperature and cold gas efficiency. Namely the higher heat capacity of CO_2 compared to N_2 is seen as the key difference.

With the initial results, the plant was modified and new gas equipment was ordered, commissioned, calibrated and integrated. Following a short proof-of-concept test campaign, the primary result were generated over the course of three days in which air, 21v% and 25v% O_2-in-CO_2 were tested. Detailed gas analysis were performed including online and gas pipettes sampled gas composition, SPA tests for tars and gas pipette samples for sulphur. The results confirmed the main investigations of the modeling studies, as operation temperatures were seen to slightly decrease the partial oxidation temperature by 52-69^{o}C and grate temperature by 31-36^{o}C. Tests with 25v% oxygen were also carried out with slightly higher temperatures. Detailed gas analysis showed that N_2 had effectively been reduced to a few percent and that tar and sulphur levels were similar to the very high standards of the air-blown operation: only a few mg/Nm^3 of tar and <3 ppm sulphur were detected. It is expected that the remaining N_2 can be completely removed by replacing the N_2-blown filter flushing system. The lone gas cleaning, a bag filter, was found to be virtually inactive for capturing these impurities.

Upscaling potentials for the TwoStage gasifier

With a foothold in a wide knowledgebase of the biomass gasification subprocesses, state-of-the-art plants and previous TwoStage upscaling developments, this project investigated the large-scale and flexibility potentials of the TwoStage system.

The project analysed the potential for two fixed beds and two fluid bed systems with wood in Article IV. This study aimed at implementing novel solutions and provide a foundation for further developmental work through comparative analysis between the two reactor types. Two noteworthy new design features were implemented: 1) applying a pyrolysis reactor with recirculation of gas, proved to be an efficient method for utilizing available heat sources indirectly and avoid dilution, which enabled excellent heat integration and high-temperature POX respectively; 2) applying subsequent cooling of the hot POX gases to match the gasification reactor with drying steam, either via direct injection or ejector-driven gas recirculation, proved to be an efficient method for enabling high POX temperatures (with high tar conversion). These two features led to three design concepts, which were shown to achieve very high cold gas efficiencies and are expected to require minimal tar removal downstream – either without any or with a simple active carbon filter. A fourth Steam-blown fluid bed concept were designed without the novel features as a reference to the other three, applying steam-blown pyrolysis and low-temperature POX. Through a thermodynamic modelling study, the four concepts were evaluated and it was found that the design features were applicable to both fixed and fluid beds. The three concepts with the new design features achieved cold gas efficiencies of 88-93%, while the reference Steam-blown fluid bed achieved 85%. The concept with two updraft reactors proved to be especially interesting with its high efficiency of 93% and increased tolerance to fines (compared to the Downdraft concept).
While still in the developmental phase, the cold gas efficiencies were found to be 6-22%-points higher than current state-of-the-art gasifiers, whilst applying minimal or very simple gas cleaning for comparison.

Further developmental work and analysis of fuel and operational flexibility were investigated in Article V. The aim was to apply the findings in Article IV and design systems capable of polygeneration: either producing a N_2-diluted gas for SOFC operation (with an estimated tar tolerance of 2g/Nm^3) with air as gasification medium, or a N_2-free gas with a minimal tar concentration for fuel synthesis reactors (estimated tar tolerance of 0.1g/Nm^3) with 50v% O_2-H_2O. Three different plant configurations were presented: all applied fluid bed char gasification reactors, but with either an updraft, slow fluid bed or fast fluid bed pyrolysis coupling. In order to expand the analysis, the use of both wood and straw fuels were investigated for all three concepts and operational states. The three concepts were thermodynamically modeled and all three design concepts were found to be technically feasible within the framework. The results showed relatively stable operation across all of the states, with blower and steam consumptions, as well as POX temperatures were within range of each other. The cold gas efficiencies were calculated on average to 83-88% and varied within 3% across fuels and gasification media for the individual concepts. The concept featuring an updraft pyrolyzer where further discussed, as it proved most promising with regards to simplicity and cold gas efficiencies. Specifically, the use of straw were subject to parameter investigations, as it proved to be significantly more steam consuming than wood. It were suggested to lower the carbon conversion for straw by 5%-points in order to streamline the operation, which results in a 4%-point cold gas efficiency loss, but conversely might enable more stable operation and better use of the char by-product.
In line with Article IV, the design were found to be technically competitive with current state-of-the-art gasifier with regards to cold gas efficiency and gas cleaning equipment, but also fuel flexibility.

Overall conclusions

- It is technically feasible to couple the TwoStage Viking gasifier with an SOFC and achieve high electric efficiencies without extensive additions of equipment or any change in gasifier operation.

The system provided state-of-the-art results and showcases the high potential of the gasifier-SOFC coupling. The cells are likely tolerant to smaller concentrations of sulphur, but it is recommended to apply some level of desulphurization (and for other species if needed) to protect the costly component. Gasifier design can however greatly simplify the level of gas cleaning. For power production the system will likely need a combined cycle power system downstream the stack in order to be competitive with a simple engine configuration, as the increase of ≈5% efficiency might not be sufficient to justify the increased complexity in comparison.

- The study has shown that the Viking plant has ability to apply alternating operation with air and O_2-CO_2 mixtures as gasification media without major changes in operation. Hence, a proof-of-concept for integrating TwoStage gasification in the polygeneration context has been provided. The system was only slightly modified and provided very high gas quality in-line with air-blown operation. Adjusting the O_2:CO_2 ratio can further optimize operation and provide higher efficiencies, less diluted product gas and enable higher POX temperatures if needed.

- From a thermodynamic point-of-view, it is possible to design very effective, large-scale TwoStage gasifier systems within the boundaries of the current technical literature. Designs for polygeneration were also found to be efficient and competitive. All of these designs are technically competitive to current state-of-the-art systems on key areas, which are central to the feasibility of the plants. As a concrete outcome, an updraft gasifier concept has been formulated as a patent application and is currently being constructed for the EP2GAS research project at DTU (EUDP no. 64017-0011).

- This study has provided the academic foundation for thermodynamically developing and/or evaluating gasifier systems going forward. Results across reactor platforms as well as fuel types and gasification media, has provided a multi-dimensional analysis of strengths and weaknesses, as well as discussions, for a given reactor constellation in a two-stage system. Hence, future opportunities and assessments can be engaged from an improved starting point, as these systems will be become increasingly relevant for the entire energy system going forward.

8.2 Further work

SOFC operation on tarry gas

Several studies have investigated the effect of tars in the feed gas to the SOFC with an overall indication that this is technically feasible to some extent. As gas cleaning equipment aimed at tars typically are a significant part of the economics of gasification systems, it could be interesting to explore this further in order to reduce costs and explore potential synergies. Experimental tests related to this study with sulphur-cleaned pyrolysis gas have already been planned in the EP2GAS project at DTU. The project will also built on the experiences and results from this thesis, and investigate solid oxide cell-integrated TwoStage gasifiers in a more complex polygeneration framework.

Further tests with oxygen-blown operation of the Viking plant

As an extension of the tests in this thesis, it would be of interest to further investigate the use of oxygen-media in the Viking gasifier plant. Specifically it could be interesting to apply a higher concentration of O_2 and apply steam as the carrier gas for a product gas composition more suited for synthesis – the studied

H_2:CO ratio is ≈1:1 in both tests and modelling. Note that the lack of H_2 may be offset by the electrolyser. As seen in Article III, the efficiency of the gasifier is expected to increase parallel to the oxygen fraction, and the handling of the carrier gas would also be simpler in smaller flows. Specifically, it is suggested to test 50v% O_2-CO_2 and O_2-H_2O, which is a trade-off between the partial oxidation temperature, efficiency, handling and operation of the solid oxide electrolysis cell. Such a study would serve multiple purposes: 1) enable direct comparison of the two carrier gases and expose/clarify differences in operation and handling; 2) enable extended proof-of-concept tests for the TwoStage-SOEC coupling; 3) investigate the application of partially oxidizing pyrolysis gas for synthesis applications.

Case-specific development of a TwoStage fluid bed gasifier model

The modelling studies presented here has been vague with regards to specific applications, as the key focus has been on the gasifier. As a next step it would however be beneficial to consider more relevant cases in order to specify requirements for e.g. fuel capacity, potential drying systems, ash sintering temperatures, H_2:CO ratios for fuel synthesis. As mentioned in Article V, it could be especially relevant to consider cases where alternative/low-grade fuels could be utilized in order to improve the plant economics. And while fuel flexible designs has been presented, systems focused on low-temperature gasification could be worth investigating. Cases for studies could be:

1) Conversion of agricultural wastes, straw and local residues with low ash-sintering temperatures, into biochar and biofuel. In an agricultural system this study could also feature integration with anaerobic digestion and synthesis of ammonia as fertilizer. Hence a more sustainable system with a high recovery of nutrients and carbon could be designed around the gasifier.
2) Integration of a gasification system in waste systems in which there is a need to address hazardous materials and utilize circular economy concepts. This could be the thermal conversion of industrial waste fractions for recovery of either high-value ash fractions (e.g. precious metals or phosphor) and/or hazardous heavy metals.

8.3 Outlook for polygeneration

Going towards 2030 and 2050, it is becoming clear through a series of analysis from energy entities that the future Danish energy system will be challenged in several ways as we progress towards non-fossil energy. In all cases, a relatively large share of bioenergy is projected [13][63].

Some of the key challenges for the Danish system that relates to this project include:

- **Costs**: Reorganizing and modifying the energy infrastructure will come at a cost, due to investments, research and increasing fuel prices etc. In order to obtain the most economically feasible transition towards an increasingly higher degree of electrification via fluctuating wind and solar power, increased use of bioenergy is central. This is mainly because the existing infrastructure can be directly used or retrofitted via to handle biomass as a substitute for fossil fuels via e.g. boiler conversions from coal to biomass, storage of bio-methane in the natural gas grid or modifications of internal combustion engines for liquid biofuels. The accessible biomass potential is mainly bound in lignocellulosic wood and straw [63], which will require thermal conversion in order to fully utilize the potential. The biomass consumption is, because of its feasibility, generally predicted to match the national potential, which could make electrolyzer integration essential for boosting (doubling) the biofuel yield/potential.

- **Transport fuels**: While it is commonly agreed upon that electrification of the passenger car fleet is the most feasible, the general projection is that heavy road, naval and air transport will be dominated by biofuels [13][63]. While there is a clear lack of political decisiveness, the most cost-effective fuels will likely be a catalogue of liquid fuels (methanol, DME etc.) and bio-methane which is/can be easily integrated in the infrastructure. It is worth noting that The Danish Energy Agency predicts that more than 50% of the transition costs towards a fossil-free energy system will come from the transport sector, which makes cost-effective solutions vital [63].
- **Storage solutions and grid balancing**: The commissioning of large capacities of wind and solar power will likely require extensive storage solutions in order handle the increasingly large fluctuations on the power grid. Batteries are most likely to be feasible in day-to-day storage, while transmission systems are limited geographically and will be pressured in the North Sea area, where several countries are expanding their wind power capacities significantly [13]. Therefore power-to-gas applications, such as electrolyzers, might end up being vital storage components, which will enable very large shares of cheap wind and solar power in the grid. Hence, seasonal storage might be enabled if the resulting electrolytic hydrogen is integrated into the energy system via biofuels. Alternately, the system should feature a smaller share of wind and solar power, and be accompanied by a large share of flexible biomass-based power plants.

For the reasons above, biomass gasification, with or without electrolyzer-integration, is expected to have a featured role in the energy system. The ability to convert a wide range of feedstock into a variety of products at a high efficiency will become increasingly important. This is projected by the Danish TSO Energinet, which suggests that energy plants based on gasification and biogas coupled to SOEC will have a total electric (electrolyzer) capacity[4] in the range of 400-500MW_e in 2-5 areas in 2035 (average of three scenarios) [13]. It is predicted that by 2035 power-to-gas will represent less than 10% of the total electricity consumption. The study also investigates polygeneration and concludes that such plants will likely produce peak-load power 11-23% of the time and generally have a viable positive economic impact on the national grid. As a part of overall research project for this study (ForskVE-12205), Sigurjonsson & Clausen [9] also investigated a polygeneration system[5] via modeling and techno-economic analysis. They found that the implementation of polygeneration would enable both a larger plant capacity factor and higher overall economic feasibility. The study also found that future prices of power, heat and bio-methane greatly influenced the operational state. Without accounting for possible subsidies in 2035, the study found that varying the bio-methane price from high (76€/MWh) to low (46€/MWh) will result in 52% and 92% of the operating time in electricity mode respectively.

The future of polygeneration will, from a technical standpoint, be dependent on the continued evolution of the gasifier and solid oxide cell technologies, and the joint platform of the two. Biomass gasification is a mature technology in the sense that it has numerous plant references and there are practical, tested solutions to most challenges. The development is however limited with regards to fuel synthesis reference plant in the MW_{th}-range, which sharply increases the cost and risk. In order to improve the feasibility of such plants, technical developments should aim at systems that can process low-grade or waste fuels, and allow integration into circular systems e.g. agricultural use of nutrient-rich biochar, power-balancing

[4] which is in the range of the gasifier input fuel capacity in such plants [7][86]
[5] based on TwoStage gasification, solid oxide electrolysis and bio-methane

polygeneration concepts and flexible energy plants for synthesis fuels and power production. Solid oxide cells still need further maturation in order to be competitive, which is generally associated with the current high investment costs. The joint gasifier-SOC platform has been subject to theoretical investigations and experimental steady-state proof-of-concepts, and the next logical step would be to analyze and operate the platform in a dynamic environment.

This study has hopefully contributed to further develop the gasification technology, deliver relevant proof-of-concepts and provide an analytical gasifier-centric view on polygeneration systems. It has been the thesis aim to expand our current understanding of polygeneration systems and assist in achieving the most efficient transition towards a sustainable energy future.

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